

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

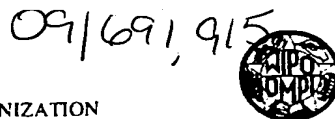
IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

THIS PAGE BLANK (USPTO)

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : A01N 43/80 // (A01N 43/80, 37:40)	A1	(11) International Publication Number: WO 97/22253 (43) International Publication Date: 26 June 1997 (26.06.97)
(21) International Application Number: PCT/EP96/05697 (22) International Filing Date: 19 December 1996 (19.12.96) (30) Priority Data: 60/008,912 20 December 1995 (20.12.95) US (71) Applicant: RHONE-POULENC AGRICULTURE LIMITED [GB/GB]; Fyfield Road, Ongar, Essex CM5 0HW (GB). (72) Inventors: ANDERSON-TAYLOR, Gordon; Rhone-Poulenc Agriculture Limited, Fyfield Road, Ongar, Essex CM5 0HW (GB). IRONS, Stephen, Malcolm; 7941 Zachary Circle, Lincoln, NE 68507 (US). LUSCOMBE, Brian, Malcolm; Rhone-Poulenc Agriculture Limited, Fyfield Road, Ongar, Essex CM5 0HW (GB). GAMBLIN, Alan; Rhone-Poulenc Agriculture Limited, Fyfield Road, Ongar, Essex CM5 0HW (GB). (74) Agent: BRACHOTTE, Charles; Rhône-Poulenc Agrochimie, Boîte postale 9163, F-69263 Lyon Cédex 09 (FR).		(81) Designated States: AL, AU, BA, BB, BG, BR, CA, CN, CU, CZ, EE, GE, HU, IL, IS, JP, KP, KR, LC, LK, LR, LT, LV, MG, MK, MN, MX, NO, NZ, PL, RO, SG, SI, SK, TR, TT, UA, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG). Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: HERBICIDAL COMPOSITIONS CONTAINING 4-BENZOYLISOXAZOLES AND A HYDROXYBENZONITRILE (57) Abstract <p>The invention relates to a composition comprising (a) 4-benzoylisoazole herbicide; and (b) bromoxynil or ioxynil, or an agriculturally acceptable salt or ester thereof, or a mixture thereof; and to the use of this composition in controlling the growth of weeds.</p> <p style="text-align: center;">Q 3 +</p>		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
AU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgyzstan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	KZ	Kazakhstan	SG	Singapore
CH	Switzerland	LI	Liechtenstein	SI	Slovenia
CI	Côte d'Ivoire	LK	Sri Lanka	SK	Slovakia
CM	Cameroon	LR	Liberia	SN	Senegal
CN	China	LT	Lithuania	SZ	Swaziland
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	LV	Latvia	TG	Togo
DE	Germany	MC	Monaco	TJ	Tajikistan
DK	Denmark	MD	Republic of Moldova	TT	Trinidad and Tobago
EE	Estonia	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	UG	Uganda
FI	Finland	MN	Mongolia	US	United States of America
FR	France	MR	Mauritania	UZ	Uzbekistan
GA	Gabon			VN	Viet Nam

HEBICIDAL COMPOSITIONS CONTAINING 4-BENZOYLISOXAZOLES AND A HYDROXYBENZONITRILE

The present invention relates to a method of controlling the growth of weeds by the application of hydroxybenzonitrile herbicides such as bromoxynil or ioxynil, or agriculturally acceptable salts or esters thereof, and certain 4-benzoylisoxazoles, and to compositions containing them.

Bromoxynil [3,5-dibromo-4-hydroxybenzonitrile] and ioxynil [4-hydroxy-3,5-diiodobenzonitrile] are known as "HBN" (hydroxybenzonitrile) herbicides and may be used for post-emergence weed control in maize, wheat and barley. Although giving control of a wide range of broad-leaf weeds, control of some important species, for example Amaranthus retroflexus, Ipomoea purpurea, Stellaria media, and Viola arvensis is unreliable. Owing to a lack of residual activity in the soil HBN herbicides do not control the weeds which emerge after application. Bromoxynil and ioxynil have no useful activity against grass or sedge weeds.

It is to be understood that where in this specification reference is made to "HBN herbicides" it is intended to refer, where the context so permits, to bromoxynil or ioxynil in the form of the parent phenol (acid equivalent: a.e.), an agriculturally acceptable salt or ester thereof, preferably an agriculturally acceptable metal or amine salt, or an agriculturally acceptable ester thereof with an alkanoic acid containing from 2 to 10 carbon atoms, or to mixtures thereof.

4-Benzoylisoxazoles are known from the literature, see for example European Patent Publications Nos. 0418175, 0487357, 0527036, 0560482, 0580439, 0609797, 0609798; and WO04/14782 and WO94/18179. Also, the compound 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole has been disclosed

CONFIRMATION COPY

as giving good pre-emergence control of grasses and broad-leaved weeds in maize (Vrabel et al; Proc. N. Cert. Weed Sci. Soc., 1994,49). The use of this compound in post emergence treatments is not however discussed.

5 As a result of research and experimentation it has now been discovered that the use of certain 4-benzoylisoxazole derivatives in combination with HBN herbicides add to their capabilities of controlling a wide spectrum of broad-leaf weeds, grasses and sedges by both foliar activity and residual soil activity.

10 In addition to this it has been found that the combined herbicidal activity of combinations of 4-benzoylisoxazole derivatives with HBN herbicides against certain species is greater than expected when applied pre- or preferably post-emergence (e.g. as a post-emergence spray) i.e. the herbicidal activity of combinations of 4-benzoylisoxazole derivatives
15 with HBN herbicides, e.g. bromoxynil showed an unexpected degree of synergism, as defined either by P.M.L. Tammes, Netherlands Journal of Plant Pathology, 70 (1964), pp 73-80 in a paper entitled "Isoboles, a graphic representation of synergism in pesticides"; or by Limpel, L.E., P.H. Schuldt and D. Lamont, 1962, 1. Proc. NEWCC 16, 48-53, using
20 the formula:-

$$E = X + Y - \frac{X \cdot Y}{100}$$

100

also known as the Colby formula (Colby S.R., 1967, Weeds 15, 20-22), where:

25 E = the expected percent inhibition of growth by a mixture of two herbicides A and B at defined doses.

X = the percent inhibition of growth by herbicide A at a defined dose.

Y = the percent inhibition of growth by herbicide B at a defined dose.

When the observed percentage of inhibition by the mixture is greater than the expected value E using the formula above the combination is synergistic.

The unexpected synergistic effect gives improved reliability of control of a number of weed species and allows for a reduction in the amount of active ingredients employed.

A high level of control of these weeds is desirable to prevent:-

- 1) yield loss, through competition and/or difficulties with harvest,
- 2) crop contamination leading to storage and cleaning difficulties, and
- 3) unacceptable weed seed return to the soil.

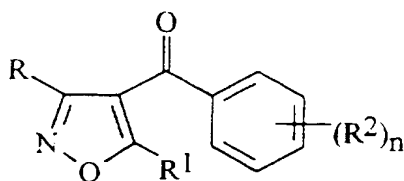
Accordingly the present invention provides a method for the control of the growth of weeds at a locus which comprises applying to the locus a herbicidally effective amount of:

- (a) a 4-benzoylisoxazole herbicide; and
- (b) bromoxynil or ioxynil or an agriculturally acceptable salt or ester thereof, preferably a metal or amine salt or an ester thereof with an alkanolic acid containing from 2 to 10 carbon atoms.

For this purpose, the 4-benzoylisoxazole herbicide and HBN herbicide are normally used in the form of herbicidal compositions (i.e. in association with compatible diluents or carriers and/or surface-active agents suitable for use in herbicidal compositions), for example as hereinafter described.

Preferably the 4-benzoylisoxazole is of formula (I):

- 4 -



(I)

wherein

R is hydrogen or -CO₂R³;

R¹ is cyclopropyl;

R² is selected from halogen (preferably chlorine or bromine),
-S(O)_pMe and C₁₋₆ alkyl or haloalkyl (preferably trifluoromethyl);

n is two or three; p is zero, one or two; and

R³ is C₁₋₄ alkyl.

In formula (I) above, compounds in which n is three and the
groups (R²)_n occupy the 2, 3 and 4-positions of the benzoyl ring; or in
which n is two and the groups (R²)_n occupy the 2- and 4- positions of
the benzoyl ring are preferred.

In formula (I) above, preferably one of the groups R² is
-S(O)_pMe.

The 4-benzoylisoxazoles of particular interest in combination with
HBN herbicides include the following:

A 5-cyclopropyl-4-(2-methylsulphonyl-4-
trifluoromethyl)benzoylisoxazole;

B 5-cyclopropyl-4-(4-methylsulphonyl-2-
trifluoromethyl)benzoylisoxazole;

C 4-(2-chloro-4-methylsulphonyl)benzoyl-
5-cyclopropylisoxazole;

D 4-(4-chloro-2-methylsulphonyl)benzoyl-
5-cyclopropylisoxazole;

E 4-(4-bromo-2-methylsulphonyl)benzoyl-
5-cyclopropylisoxazole;

F ethyl 3-[5-cyclopropyl-4-(3,4-dichloro-2-
methylsulphenyl)benzoylisoxazole]carboxylate;

5 G 5-cyclopropyl-4-(3,4-dichloro-2-
methylsulphonyl)benzoylisoxazole; and

H ethyl 3-[5-cyclopropyl-4-(2-methylsulphonyl-4-
trifluoromethyl)benzoylisoxazole]carboxylate.

10 Of these, compounds A, F and H are preferred, especially
Compounds A and F.

The HBN herbicide is preferably present as either the parent
phenol or as an ester, most preferably selected from the butyrate,
heptanoate and octanoate or mixtures thereof.

15 The amounts of the 4-benzoylisoxazole herbicide and HBN
herbicide applied vary with the nature of the weeds, the compositions
used, the time of application, the climatic and edaphic conditions and
(when used to control the growth of weeds in crop-growing areas) the
nature of the crops. When applied to a crop-growing area, the rate of
20 application should be sufficient to control the growth of weeds without
causing substantial permanent damage to the crop. In general, taking
these factors into account, application rates from 5g to 500g of the
4-benzoylisoxazole herbicide and from 30 g to 600g a.e. of HBN
herbicide per hectare give good results. However, it is to be understood
that higher or lower application rates may be used, depending upon the
25 particular problem of weed control encountered.

The 4-benzoylisoxazole herbicide and HBN herbicide in
combination may be used to control selectively the growth of weeds, for

example to control the growth of those species hereinafter mentioned, by pre- or preferably post-emergence application in a directional or non-directional fashion, e.g. by directional or non-directional spraying, to a locus of weed infestation which is an area used, or to be used, for growing crops, for example wheat, barley, oats, rye, maize and rice, soya beans, field and dwarf beans, peas, lucerne, cotton, peanuts, flax, onions, carrots, oilseed rape, sunflower, sugarcane, potatoes and permanent or sown grassland before or after sowing of the crop or before or after emergence of the crop.

For the selective control of weeds at a locus of weed infestation which is an area used, or to be used, for the growing of crops application rates from 5 g to 500 g of the 4-benzoylisoxazole and from 30g to 360g a.e. of HBN herbicide per hectare are particularly suitable, more preferably from 20 to 300 g of the 4-benzoylisoxazole herbicide and from 60 g to 200 g a.e. of HBN herbicide per hectare.

The use of the 4-benzoylisoxazole herbicide and HBN herbicide for the control of weeds in maize, winter or spring cereals, or sugarcane is preferred.

Where the 4-benzoylisoxazole is Compound A, the HBN herbicide is bromoxynil and the mixture is for use in maize, application rates of from 5 to 75 g per hectare of the 4-benzoylisoxazole and from 20 g to 280 g a.e. per hectare of bromoxynil are preferred, more preferably from 10 to 50 g of 4-benzoylisoxazole and from 35 g to 280 g a.e. of bromoxynil per hectare, even more preferably from 15 to 25 g of 4-benzoylisoxazole and about 140 g a.e. of bromoxynil per hectare.

Where the 4-benzoylisoxazole is Compound A, the HBN herbicide is bromoxynil and the mixture is for use in spring cereals, application rates of from 5 to 100 g per hectare of the 4-benzoylisoxazole herbicide

and from 30 g to 280 g a.e. per hectare of bromoxynil are preferred, more preferably from 10 to 60 g of 4-benzoylisoxazole and from 60 g to 280 g a.e. of bromoxynil per hectare, even more preferably from 20 to 40 g of 4-benzoylisoxazole and about 120 g a.e. of bromoxynil per hectare.

5 Where the 4-benzoylisoxazole is Compound A, the HBN herbicide is bromoxynil and the mixture is for use in winter cereals, application rates of from 10 to 150 g per hectare of the 4-benzoylisoxazole and from 60 g to 360 g a.e. per hectare of bromoxynil are preferred, more preferably from 10 to 60 g of 4-benzoylisoxazole and from 80 g to 280 g
10 a.e. of bromoxynil per hectare, even more preferably from 20 to 40 g of 4-benzoylisoxazole and about 120 g a.e. of bromoxynil per hectare.

 Where the 4-benzoylisoxazole is Compound F and the HBN herbicide is bromoxynil the mixture is most preferably used in controlling
weeds found in maize. In this use application rates of from 25 to 300 g
15 per hectare of the 4-benzoylisoxazole and from 60 to 120 g a.e. per hectare of bromoxynil are preferred, more preferably from 150 to 250 g per hectare of the 4-benzoylisoxazole herbicide and about 120 g a.e. of bromoxynil.

 Where the HBN herbicide is ioxynil the mixture is preferably used
20 to control weeds found in cereal crops such as wheat and barley (in particular winter wheat and winter barley). The ioxynil is preferably used in the form of an ester, preferably the octanoate ester.

 The mixtures of the present invention may also be used to control the growth of weeds at loci which are not crop-growing areas but in
25 which the control of weeds is nevertheless desirable. Examples of such non-crop-growing areas include airfields, industrial sites, railways, roadside verges, the verges of rivers, irrigation and other waterways, scrublands and fallow or uncultivated land, in particular where it is

desired to control the growth of weeds in order to reduce fire risks

When used for such purposes in which a total herbicidal effect is frequently desired, the active compounds are normally applied at dosage rates higher than those used in crop-growing areas as hereinbefore described. The precise dosage will depend upon the nature of the vegetation treated and the effect sought. In general, application rates of from 125 g to 500 g per hectare of the 4-benzoylisoxazole herbicide and from 30 g to 600 g a.e. per hectare of HBN herbicide are used, more preferably from 150 g to 250 g of 4-benzoylisoxazole herbicide and from 60 to 280 g a.e. per hectare of HBN herbicide.

The following non-limiting examples illustrate the method of the invention. Unless otherwise specified, in the examples the application rates for the HBN herbicides refer to the amount of active ingredient present. In the description that follows the following Bayer codes are used for the various weed and crop species:

Bayer Code	Species	Bayer Code	Species
GALAP	<u>Galium aparine</u>	ABUTH	<u>Abutilon theophrasti</u>
VERHE	<u>Veronica hederifolia</u>	AMARE	<u>Amaranthus retroflexus</u>
VIOAR	<u>Viola arvensis</u>	AMATA	<u>Amaranthus sp</u>
STEME	<u>Stellaria media</u>	SETFA	<u>Setaria faberi</u>
MATCH	<u>Matricaria chamomilla</u>	SETLU	<u>Setaria lutescens</u>
FUMOF	<u>Fumaria officinalis</u>	SORVU	<u>Sorghum vulgare</u>
MATSS	<u>Matricaria spp.</u>	SORHA	<u>Sorghum halepense</u>
PAPRH	<u>Papaver rhoeas</u>	CASOB	<u>Cassia obtusifolia</u>
SINAR	<u>Sinapis arvensis</u>	SIDSP	<u>Sida spinosa</u>
PANI	<u>Panicum dichotomiflorum</u>	LAMAM	<u>Lamium amplexicaule</u>
		TRAW	<u>Triticum aestivum</u>

Example 1

The following field trials were conducted post-emergence of the weed and crop species at a research farm in Nebraska, U.S.A. (a silt loam soil; pH 7.0; content 18% sand; 56% silt and 26% clay).

5 Compound A (formulated as a wettable powder) and bromoxynil (as the commercially available emulsifiable concentrate "Buctril 2EC", trademark the octanoate ester) were dissolved in water and applied either alone or in mixtures to approximately 7 square metre plots at a spray volume rate of from 100 to 300 litres/ha. The crop and weed species
10 had been drill sown 27 days earlier. Three replicates were performed and the percentage phytotoxicity was assessed visually 35 days after treatment in comparison with an untreated control.

In the tables that follow, the figure in brackets represents the expected percentage level of control according to the Colby formula.

15

RESULTS**Table A1****Setaria faberi**

Compound A	Bromoxynil		
	Dose (g/ha)	0	210
	0	-	17
	12.5	20	65(34)
	25	40	89(50)
	50	77	98(81)

Table A2**Digitaria sanguinalis**

Compound A	Bromoxynil		
	Dose (g/ha)	0	210
	0	-	20
	12.5	20	83(36)
	25	63	100(70)
	50	93	100(94)

20

Table A3**Setaria viridis**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	27
	12.5	27	62(47)
	25	40	87(56)
	50	78	88(84)

Table A4**Panicum dichotomiflorum**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	23
	12.5	30	68(46)
	25	50	100(62)
	50	78	100(83)

5

Table A5**Echinochloa crus-galli**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	17
	12.5	33	93(44)
	25	78	100(82)
	50	100	100(100)

Table A6**Sorghum vulgare**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	16
	12.5	27	57(39)
	25	40	90(50)
	50	78	98(82)

Table A7**Setaria glauca**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	17
	12.5	27	53(39)
	25	38	88(49)
	50	73	88(78)

Table A8**Maize (Zea Mays; variety: PIONEER 3394)**

		Bromoxynil	
Compound A	Dose (g/ha)	0	210
	0	-	0
	12.5	0	0(0)
	25	0	5(0)
	50	0	8(0)

5

Example 2

The following glasshouse experiments were conducted to determine the efficacy of Compound F with bromoxynil as described below.

10 Compound F was formulated as a 50% wettable powder containing the following ingredients:

	Compound F	50%
	Arylan SX flake	3%
	Arkopon T	5%
15	Sopropon T36	1%
	Tixosil 38	3%
	China Clay	38%

It was dissolved in water in the presence of 0.1% 'Agral' (trademark, an alkyl phenol ethoxylate) and applied post-emergence on
 20 the weed species. Bromoxynil (as the commercial formulation 'Pardner',

trademark, at 225g/l) was dissolved in water, also in the presence of 0.1% 'Agral' and applied in the same manner at various application rates. Additionally the combination was tank mixed. The spray jet and pressure used to apply the compounds gave a volume equivalent to 290 l/ha.

5 Treatment effects were assessed visually about 14 days after treatment. In addition 'Agral' (trademark) was applied on its own to determine its intrinsic phytotoxicity. The weeds were at the following growth stage when treated:-

10	<u>Code</u>	<u>Species</u>	<u>Stage at treatment</u>
	CASOB	: <u>Cassia obtusifolia</u>	cots - 1 leaf
	SIDSP	: <u>Sida spinosa</u>	cots - 2 leaf
	CYPES	: <u>Cyperus esculentus</u>	4-5 leaf
	DIGSA	: <u>Digitaria sanguinalis</u>	2-3 leaf
15	ECHCG	: <u>Echinochloa crus-galli</u>	3 leaf
	PANDI	: <u>Panicum dichotomiflorum</u>	3 leaf
	SETFA	: <u>Setaria faberi</u>	2-3 leaf
	SETVI	: <u>Setaria viridis</u>	3 leaf
	SORHA	: <u>Sorghum halepense</u>	3 leaf

20 In the Tables that follow, "Brom" means bromoxynil as described above. The figures in brackets indicate the expected level of weed control according to the Colby formula.

Table B1

Compound	Rate (g/ha)	CASOB	SIDSP	CYPES	DIGSA	ECHCG	PANDI	SETFA	SETVI	SORHA
F (+ 0.1% Agral)	16	50	50	0	40	90	30	80	0	80
	32	70	60	20	50	95	50	90	30	95
	63	80	70	20	70	95	60	100	60	100
	125	85	70	30	85	100	90	100	80	100
Bromoxynil (+ 0.1% Agral) (BROM)	63	0	50	0	0	0	0	0	10	0
	125	30	60	0	20	20	0	10	30	0
	250	40	60	0	30	20	0	30	30	20

Table B1 (continued)

Compound	Rate (g/ha)	CASOB	SIDSP	CYPES	DIGSA	ECHCG	PANDI	SETFA	SETVI	SORHA
F + BROM	16+63	85(50)	80(75)	10(0)	60(52)	100(90)	60(30)	90(80)	30(10)	95(80)
F + BROM	32+63	100(70)	100(80)	20(20)	60(60)	100(95)	70(50)	90(90)	70(37)	95(95)
F + BROM	63+63	90(80)	90(85)	40(20)	80(76)	100(95)	70(60)	100(100)	80(64)	100(100)
F + BROM	125+63	90(85)	90(85)	40(30)	85(88)	100(100)	85(90)	100(100)	100(82)	100(100)
F + BROM	16+125	90(65)	85(80)	20(0)	60(58)	100(92)	20(30)	100(82)	60(30)	100(80)
F + BROM	32+125	80(79)	100(84)	20(20)	70(65)	100(96)	70(50)	100(91)	70(51)	100(95)
F + BROM	63+125	70(86)	90(88)	30(20)	85(79)	100(96)	90(60)	100(100)	100(72)	100(100)
F + BROM	125+125	70(90)	90(88)	50(30)	100(90)	100(100)	90(90)	100(100)	100(86)	100(100)
F + BROM	16+250	80(70)	80(80)	10(0)	60(64)	100(92)	50(30)	100(86)	80(30)	95(84)
F + BROM	32+250	80(82)	100(84)	30(20)	80(70)	100(96)	80(50)	100(93)	95(51)	100(96)
F + BROM	63+250	90(88)	100(88)	40(20)	100(82)	100(96)	90(60)	100(100)	95(72)	100(100)
F + BROM	125+250	70(91)	100(88)	70(30)	100(91)	100(100)	90(90)	100(100)	95(86)	100(100)

0.1% Agrav	-	0	0	0	0	0	0	0	0	0
------------	---	---	---	---	---	---	---	---	---	---

Example 3

The following field trial was conducted to establish the efficacy of the mixtures of the invention in winter cereals. The trial was conducted in New Jersey post emergence of the weed and crop species. Compound A (formulated as a 75% wettable granule) and bromoxynil (commercial emulsifiable concentrate formulation "Buctril", trademark containing 240g/l of the octanoate ester) were dissolved in water and applied either alone or in tank mixes of various application rates to 3 metres by 5 metres plots (sandy loam soil) in which had been sown six months earlier the weed and crop species. The spray rate was 300 l/ha and 3 replicates were performed. The phytotoxicity was assessed 35 days after treatment and at the time of treatment the growth stage of the plants was as follows:-

<u>Code</u>	<u>Species</u>	<u>Growth Stage (Height)</u>
LAMAM	<u>Lamium amplexicaule</u>	10-15 cm
STEME	<u>Stellaria media</u>	18-20 cm
TRAW	<u>Triticum aestivum</u> (Variety: Pioneer 2548)	20-25 cm

The results are shown in the table that follows (N/D means no data available; "Brom" refers to the bromoxynil described above).

Results:**TABLE C1**

Compound	Rate (g/ha)	WEED		CROP
		LAMAM	STEME	TRAW
A	10	55	65	0
A	20	47	47	0
A	40	78	83	0

Brom	60	23	20	0
Brom	120	17	23	0

A+Brom	10+60	83(65)	82(72)	0
A+Brom	10+120	68(63)	75(73)	0
A+Brom	20+60	80(59)	78(58)	0
A+Brom	20+120	93(56)	88(59)	0
A+Brom	40+60	88(83)	78(86)	0
A+Brom	40+120	93(82)	92(87)	0

Example 4

5

The following glasshouse experiments were conducted to determine the efficacy of mixtures of compound A and bromoxynil.

10

Compound A (technical material) and bromoxynil (octanoate ester, technical material) were dissolved in acetone and made up in water. Treatments were applied of the compounds alone and in tank mixes. The test solutions were applied post emergence to the plants at a spray rate of 290 litres/ha. Four replicates were

performed and the pots were placed in the glasshouse in randomised pots. They were watered overhead 24 hours after treatment and mat watered thereafter. After treatment the plants were arranged in the glasshouse in a randomised block design. Visual assessment of percentage reduction in green area was made 17 days after treatment in comparison with untreated plants.

The growth stage of the plants when treated was as follows:-

Bayer Code	Species	Growth Stage at Treatment
GALAP	<u>Galium aparine</u>	2 whorls. branching
VERHE	<u>Veronica hederifolia</u>	2-4 leaves
VIOAR	<u>Viola arvensis</u>	2 leaves

The levels of control observed for each mixture treatment were compared with expected values calculated from the response of plants to each component compound applied alone, (Colby 1967). Where possible effective dose (ED) rates giving 50% or 90% control were calculated using log/probit analysis (in the case of Galium aparine ED50 values were calculated; in the cases of Veronica hederifolia and Viola arvensis ED90 values were calculated). Isoboles were then constructed to illustrate the nature of the interaction (Tammes 1964). The expected values are shown in brackets in the Table that follows, as are the respective ED50 and ED90 values. In each case the rate is expressed in g/ha:-

Table D1

Compound	Rate	GALAP	VERHE	VIOAR
A	0.5	3	15	0
A	1	8	11	10
A	2	8	16	23
A	4	13	45	25
A	8	33	60	53
A	16	55	74	48
A	31	63	90	58
A	63	73	99	85
A	125	80	98	95
ED50/90		20	25	83
Brom	8	25	6	35
Brom	16	10	11	33
Brom	31	43	18	35
Brom	63	58	13	60
Brom	125	70	43	65
Brom	250	90	91	95
ED50/90		67	283	214
A + Brom	0.5 + 8	5 (27)	9 (20)	45 (35)
A + Brom	1 + 8	8 (31)	24 (16)	34 (42)
A + Brom	2 + 8	28 (31)	55 (21)	63 (50)
A + Brom	4 + 8	33 (35)	58 (48)	63 (51)
A + Brom	8 + 8	28 (50)	78 (62)	85 (69)
A + Brom	16 + 8	43 (66)	90 (76)	95 (66)
A + Brom	31 + 8	70 (72)	96 (91)	96 (82)
A + Brom	63 + 8	70 (80)	98 (99)	100 (90)
	ED50/90	15	9	12

Table D1 (continued)

Compound	Rate	GALAP	VERHE	VIOAR
A + Brom	0.5 + 16	13 (13)	53 (24)	50 (33)
A + Brom	1 + 16	18 (17)	70 (21)	48 (40)
A + Brom	2 + 16	38 (17)	78 (25)	63 (48)
A + Brom	4 + 16	35 (22)	83 (51)	68 (50)
A + Brom	8 + 16	58 (40)	90 (64)	96 (69)
A + Brom	16 + 16	65 (60)	96 (77)	100 (65)
A + Brom	31 + 16	70 (67)	95 (91)	99 (82)
A + Brom	63 + 16	80 (76)	95 (99)	100 (90)
	ED50/90	7	9	5

A + Brom	0.5 + 31	30 (45)	65 (30)	65 (35)
A + Brom	1 + 31	45 (48)	70 (27)	75 (42)
A + Brom	2 + 31	48 (48)	78 (31)	89 (50)
A + Brom	4 + 31	60 (50)	89 (55)	93 (51)
A + Brom	8 + 31	60 (62)	90 (67)	99 (69)
A + Brom	16 + 31	73 (74)	91 (79)	100 (66)
A + Brom	31 + 31	83 (79)	95 (92)	99 (82)
A + Brom	63 + 31	83 (85)	96 (99)	98 (90)
	ED50/90	2	17	5

A + Brom	0.5 + 63	50 (59)	80 (26)	88 (60)
A + Brom	1 + 63	63 (61)	83 (23)	78 (64)
A + Brom	2 + 63	63 (61)	88 (27)	90 (69)
A + Brom	4 + 63	70 (63)	78 (52)	98 (70)
A + Brom	8 + 63	83 (72)	90 (65)	99 (81)
A + Brom	16 + 63	83 (81)	96 (77)	100 (79)
A + Brom	31 + 63	91 (84)	96 (91)	100 (89)
A + Brom	63 + 63	95 (89)	99 (99)	100 (94)
	ED50/90	1	5	2

Brief Description of Drawings

5 Figure I is an ED50 plot calculated from observed values (---) and a corresponding plot of expected additive values (dashed line) for the mixtures of compound A with bromoxynil against the weed species Galium aparine.

Figure II is an ED90 plot calculated from observed values (---) and a corresponding plot of expected additive values (dashed line) for the mixtures of compound A with bromoxynil against the weed species Veronica hederifolia.

10 Figure III is an ED90 plot calculated from observed values (---) and a corresponding plot of expected additive values (dashed line) for the mixtures of compound A with bromoxynil against the weed species Viola arvensis.

15 The isoboles produced from the data in Table D1, shown in Figs. I to III hereafter, were clearly type III curves (Tammes, op. cit., Page 75, Fig 2), characteristic of synergism.

Example 5

The following field trials were conducted in Mereville, France (referred to below as location FR1; loamy-clay-sand soil); Seville, Spain (referred to below as location ES1; sandy-loam soil); Essex, England (referred to below as location UK1; sandy-loam soil); and Alzonne, France (referred to below as location FR2; clay-loam soil). Compound A (as a 75% by weight wettable granule) and ioxynil (as the octanoate ester, an emulsifiable concentrate sold as "Totril", trade mark) or bromoxynil (as the octanoate ester, formulated as a 20% wettable powder) were applied either alone or in tank mix combination in the early spring post emergence to winter-germinating weed species, and the percentage phytotoxicity in each weed species was assessed by comparison with an untreated control 48 days after treatment (DAT) at location FR1; 55 DAT at location ES1; 56 DAT at location UK1; and 77 DAT at location FR2.

The results were as follows with the figures in parenthesis indicating the expected control according to the Colby formula (note 'A.I.' means active ingredient; 'Brom' means bromoxynil as described above).

Results
TABLE E1

	Weed Species									
	Location	FR1	FR1	ES1	ES1	ES1	ES1	UK1	UK1	UK1
A.I.	Dose (g/ha)	STEME	VERHE	MATCH	STEME	FUMOF	STEME	VERHE		
Cpd A	22.5	83	80	0	20	20	0	0		
Brom	120	90	30	20	0	0	0	0		
Cpd A + Brom	22.5 + 120	90(98)	90(86)	15(20)	40(20)	55(20)	37(0)	53(0)		
Ioxynil	120	70	60	30	20	35	0	0		
Cpd A + Ioxynil	22.5 + 120	98(95)	100(92)	35(30)	25(36)	40(48)	53(0)	53(0)		

TABLE E1 (continued)

	Location	UK1	Weed Species							
			FR2	FR2	FR2	FR2	FR2	FR2	FR2	FR2
A.I.	Dose (g/ha)	MATSS	STEME	VERHE	MATCH	PAPRH	SINAR	VERJIE	GALAP	
Cpd A	22.5	0	15	30	40	15	65	25	5	
Brom	120	83	35	25	90	50	80	40	23	
Cpd A + Brom	22.5 + 120	43(83)	35(45)	35(48)	93(94)	45(58)	97(93)	40(55)	10(27)	
Ioxynil	120	0	45	33	93	35	92	40	38	
Cpd A + Ioxynil	22.5 + 120	37(0)	60(53)	60(53)	93(96)	45(45)	97(97)	94(55)	50(41)	

Example 6

The following field trials were conducted in Volga, USA (referred to below as location US1; silt-loam soil); and Indiana, USA (referred to below as location US2; silt-loam soil); Compound A (formulated as a 75% wettable granule) and H (formulated as a 50% wettable powder) and bromoxynil (octanoate ester, formulated as a 20% wettable powder) were applied either alone or as a tank mix early-post emergence to spring-germinating weed species in a spring-sown crop, and the percentage phytotoxicity in each weed species was compared by comparison with an untreated control 35 or 36 days after treatment. The results are shown in the Table F1 below, with the figures in parenthesis indicating the expected control according to the Colby formula (note 'A.I.' means active ingredient; 'Brom' means bromoxynil as described above).

TABLE F1

	Location	US1	US2	US1	US2	US1	US2	US1	US2
A.I.	Dose (g/ha)	ABUTH	ABUTH	AMARE	AMARE	AMATA	ECHCG	ECHCG	ECHCG
Brom	60	10	0	8	28	10	0	0	0
Brom	120	73	85	60	93	53	0	0	20
Cpd A	15	65	25	30	0	58	43	45	45
Brom + Cpd A	60 + 15	100(69)	100(25)	100(36)	100(28)	97(62)	97(43)	55(45)	55(45)
Brom + Cpd A	120 + 15	100(91)	100(89)	90(72)	98(93)	91(80)	98(43)	95(56)	95(56)
Cpd H	25	43	92	10	0	8	0	25	25
Brom + Cpd H	60 + 25	100(49)	100(92)	99(17)	100(28)	99(17)	93(0)	90(25)	90(25)
Brom + Cpd H	120 + 25	100(43)	100(91)	100(10)	100(0)	99(8)	92(0)	95(25)	95(25)

TABLE F1 (continued)

	Location	US1	US2	US1	US2	US1	US2	US1	US2	US1	US2
A.I.	Dose (g/ha)	HELAN	PANMI	SETFA	SETFA	SETLU	SETLU	SORVU	SORVU	SORVU	SORVU
Brom	60	18	0	0	0	0	0	0	0	0	0
Brom	120	73	0	0	0	0	0	0	0	0	0
Cpd A	15	30	0	0	0	28	0	10	0	0	0
Brom + Cpd A	60 + 15	100(43)	10(0)	10(0)	35(0)	67(28)	0(0)	0(10)	0(0)	0(0)	0(0)
Brom + Cpd A	120 + 15	65(81)	0(0)	53(0)	5(0)	95(28)	0(0)	23(10)	0(0)	0(0)	0(0)
Cpd H	25	20	0	0	20	0	13	8	0	0	0
Brom + Cpd H	60 + 25	85(34)	0(0)	15(0)	50(20)	65(0)	0(13)	23(8)	0(0)	0(0)	0(0)
Brom + Cpd H	120 + 25	80(20)	0(0)	5(0)	60(20)	71(0)	18(13)	0(8)	0(0)	0(0)	0(0)

According to a further feature of the present invention, there are provided herbicidal compositions comprising:

- (a) a 4-benzoylisoxazole herbicide; and
- 5 (b) bromoxynil or ioxynil, an agriculturally acceptable salt or ester thereof or a mixture thereof, preferably a metal or amine salt or an ester thereof with an alkanolic acid containing from 2 to 10 carbon atoms;

10 in association with, and preferably homogeneously dispersed in, one or more compatible herbicidally- acceptable diluents or carriers and/or surface-active agents (i.e. diluents or carriers or surface-active agents of the type generally acceptable in the art as being suitable for use in herbicidal compositions and which are compatible with bromoxynil and ioxynil and 4-benzoylisoxazole herbicides). The term "homogeneously dispersed" is used to include
15 compositions in which the HBN herbicide and 4-benzoylisoxazole herbicide are dissolved in the other components. The term "herbicidal composition" is used in a broad sense to include not only compositions which are ready for use as herbicides but also
20 concentrates which must be diluted before use.

The compositions preferably comprise 4-benzoylisoxazole herbicide and HBN herbicide in proportions of from 1:120 to 16.7:1, preferably from 1:10 to 5:1 wt/wt of (a):(b).

25 Preferably, the compositions contain from 0.05 to 90% by weight of HBN herbicide and 4-benzoylisoxazole derivative(s).

The herbicidal compositions may contain both a diluent or carrier and a surface-active (e.g. wetting, dispersing, or emulsifying)

agent. Surface-active agents which may be present in herbicidal compositions of the present invention may be of the ionic or non-ionic types, for example sulphuricinoleates, products based on condensates of ethylene oxide with nonyl- or octyl-phenols, or
5 carboxylic acid esters of anhydrosorbitols which have been rendered soluble by etherification of the free hydroxy groups by condensation with ethylene oxide, alkali and alkaline earth metal salts or sulphuric acid esters and sulphonic acids such as dinonyl- and dioctyl-sodium sulphonosuccinates and alkali and alkaline earth metal salts of high
10 molecular weight sulphonic acid derivatives such as sodium and calcium lignosulphonates. Examples of suitable solid diluents or carriers are aluminium silicate, talc, calcined magnesia, kieselguhr, tricalcium phosphate, powdered cork, absorbent carbon black and clays such as kaolin and bentonite. The solid compositions (which
15 may take the form of dusts, granules or wettable powders) are preferably prepared by grinding the HBN herbicides, e.g. bromoxynil and the 4-benzoylisoxazole derivative with solid diluents or by impregnating the solid diluents or carriers with solutions of HBN herbicide and 4-benzoylisoxazole derivative in volatile
20 solvents, evaporating the solvents and, if necessary, grinding the products so as to obtain powders. Granular formulations may be prepared by absorbing the HBN herbicide and the 4-benzoylisoxazole derivative (dissolved in volatile solvents) onto the solid diluents or carriers in granular form and evaporating the
25 solvents, or by granulating compositions in powder form obtained as described above. Solid herbicidal compositions, particularly wettable powders, may contain wetting or dispersing agents (for

example of the types described above), which may also, when solid, serve as diluents or carriers.

Liquid compositions according to the invention may take the form of aqueous-organic solutions, suspensions and emulsions which may incorporate a surface-active agent. Suitable liquid diluents for incorporation in the liquid compositions include water, acetophenone, cyclohexanone, isophorone, toluene, xylene and mineral, animal and vegetable oils (and mixtures of these diluents). Surface-active agents, which may be present in the liquid compositions, may be ionic or non-ionic (for example of the types described above) and may, when liquid, also serve as diluents or carriers.

Wettable powders and liquid compositions in the form of concentrates may be diluted with water or other suitable diluents, for example mineral or vegetable oils, particularly in the case of liquid concentrates in which the diluent or carrier is an oil, to give compositions ready for use. When desired, liquid compositions of HBN herbicide and 4-benzoylisoxazole derivative may be used in the form of self-emulsifying concentrates containing the active substances dissolved in the emulsifying agents compatible with the active substances, the simple addition of water to such concentrates producing compositions ready for use.

Liquid concentrates in which the diluent or carrier is an oil may be used without further dilution using the electrostatic spray technique.

Herbicidal compositions according to the present invention may also contain, if desired, conventional adjuvants such as

adhesives, protective colloids, thickeners, penetrating agents, stabilisers, sequestering agents, anti-caking agents, colouring agents and corrosion inhibitors. These adjuvants may also serve as carriers or diluents.

5 Preferred herbicidal compositions according to the present invention are in the form of aqueous suspension concentrates; wettable powders, liquid water soluble concentrates; liquid emulsifiable suspension concentrates; granules or emulsifiable concentrates. Where compound F is present in the herbicidal
10 composition, preferably the composition is in the form of an emulsifiable concentrate.

In addition the compositions may be provided in the form of a gel. This is particularly useful where the composition is intended for packaging in a water soluble bag for example as described in
15 European Patent Publication Nos. 0577702 and 0608340, and U.S. Patent Nos. 5,222,595; 5,224,601; 5,351,831; and 5,323,906.

The processes described in European Patent Publication Nos. 0418175, 0487357, 0527036 and 0560482 may be used to prepare the compounds of formula (I).

20 Herbicidal compositions according to the present invention may also comprise the HBN herbicide and 4-benzoylisoxazole herbicide in association with, and preferably homogeneously dispersed in, one or more other pesticidally active compounds and, if desired, one or more compatible pesticidally acceptable diluents
25 or carriers, surface-active agents or conventional adjuvants as hereinbefore described.

Pesticidally active compounds and other biologically active materials which may be included in, or used in conjunction with, the herbicidal compositions of the present invention, for example those hereinbefore mentioned, and which are acids, may, if desired, be
5 utilised in the form of conventional derivatives, for example alkali metal and amine salts and esters. Preferably the additional biologically active material is a herbicide.

The following Examples illustrate compositions according to the invention. In the description that follows the following
10 trademarks appear:- Tergitol, Atlox, Aerosol OT/B, Solvesso, Arylan, Synperonic.

Example C1

The following formulation was prepared:

Compound F	20%
Bromoxynil	20%
Tergitol; XD	4%
Atlox G3300B	5%
Sodium lauryl sulphate	2%
Aerosol OT/B	0.5%
Sodium acetate	0.4%
Solvesso 200	to 100%

This was processed to subsequently yield a gel formulation
15 according to methods known in the art.

Example C2

An emulsifiable concentrate was prepared using the following ingredients:

Compound F	20%
Bromoxynil	45%
Arylan CA	4%
Synperonic NPE1800	4%
Solvesso 200	to 100%

5

by dissolving the active ingredients in Solvesso 200 solvent at 50°C. The mixture was cooled, the remaining components were then added and the formulation was made up to volume with Solvesso 200 solvent.

10

By proceeding in a similar manner the following emulsifiable concentrates were prepared:

Example C3

Compound F	20%
Bromoxynil	10%
Arylan CA	4%
Synperonic NPE1800	4%
Solvesso 200	to 100%

Example C4

Compound F	6%
Bromoxynil	42%
Arylan CA	4%
Synperonic NPE1800	4%
Solvesso 200	to 100%

15

Example C5

Compound F	6%
Bromoxynil	12%
Arylan CA	4%
Synperonic NPE 1800	4%
Solvesso 200	to 100%

According to a further feature of the present invention, there is provided a product comprising:

- 5 (a) a 4-benzoylisoazole herbicide; and
- (b) bromoxynil or ioxynil, an agriculturally acceptable salt or ester thereof or a mixture thereof, preferably a metal or amine salt or an ester thereof with an alkanolic acid containing from 2 to 10 carbon atoms;
- 10 as a combined preparation for simultaneous, separate or sequential use, for example, in controlling the growth of weeds at a locus, e.g. crop locus.

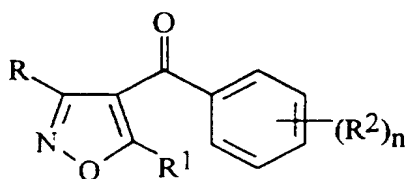
CLAIMS.

1. A method for the control of the growth of weeds at a locus which comprises applying to the locus a synergistic herbicidally effective amount of:

- 5 (a) a 4-benzoylisoxazole herbicide; and
 (b) bromoxynil, which is 3,5-dibromo-4-hydroxybenzonitrile, or ioxynil, which is 4-hydroxy-3,5-diiodobenzonitrile, an agriculturally acceptable salt or ester thereof, or a mixture thereof.

10

2. A method according to Claim 1 in which the 4-benzoylisoxazole is a compound of formula (I):



(I)

15

wherein

R is hydrogen or $-\text{CO}_2\text{R}^3$;

R^1 is cyclopropyl;

R^2 is selected from halogen, $-\text{S}(\text{O})_p\text{Me}$ and C_{1-6} alkyl or haloalkyl;

20

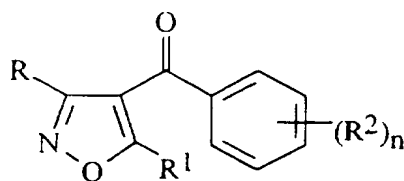
n is two or three; p is zero, one or two; and

R^3 is C_{1-4} alkyl.

25

3. A method for the control of the growth of weeds at a locus which comprises applying to the locus a synergistic herbicidally effective amount of:

(a) a 4-benzoylisoxazole of formula (I)



(I)

wherein

5

R is hydrogen or $-\text{CO}_2\text{R}^3$;

R^1 is cyclopropyl;

R^2 is selected from halogen, $-\text{S}(\text{O})_p\text{Me}$ and C_{1-6} alkyl or haloalkyl;

n is two or three; p is zero, one or two; and

10

R^3 is C_{1-4} alkyl; and

(b) bromoxynil, which is 3,5-dibromo-

4-hydroxybenzonitrile, or an agriculturally acceptable salt or ester thereof.

15

4. A method according to claim 1, 2 or 3 using an application rate of from 5 g to 500 g of (a) and from 30g to 600g acid equivalent of (b) per hectare.

20

5. A method according to any one of claims 1 to 4 using bromoxynil or ioxynil as an agriculturally acceptable metal or amine salt, or an agriculturally acceptable ester thereof with an alkanolic acid containing from 2 to 10 carbon atoms.

25

6. A method according to any one of claims 2 to 5 wherein, in formula (I), n is three and the groups $(\text{R}^2)_n$ occupy the

2, 3 and 4-positions of the benzoyl ring; or in which n is two and the groups $(R^2)_n$ occupy the 2 and 4- positions of the benzoyl ring.

7. A method according to any one of claims 2 to 6
5 wherein, in formula (I), one of the groups R^2 is $-S(O)_pMe$, wherein p is as defined in claim 2.

8. A method according to any one of claims 2 to 7
10 wherein the compound of formula (I) is 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethyl)benzoylisoxazole.

9. A method according to any one of claims 2 to 7
15 wherein the compound of formula (I) is ethyl 3-[5-cyclopropyl-4-(3,4-dichloro-2-methylsulphenyl)benzoylisoxazole]-carboxylate.

10. A method according to any one of claims 1 to 9 by post-emergence application.

11. A method according to any one of claims 1 to 10 in
20 which the locus is an area used, or to be used, for growing maize or winter cereals.

12. A composition comprising a synergistic herbicidally effective amount of:

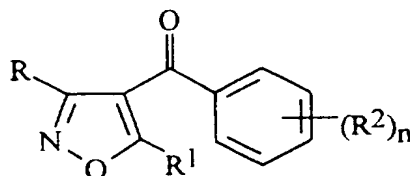
25 (a) a 4-benzoylisoxazole derivative as defined in any one of claims 1, 2 or 6 to 9; and

(b) bromoxynil, which is 3,5-dibromo-4-hydroxybenzonitrile, or ioxynil, which is 4-hydroxy-3,5-diiodobenzonitrile, an agriculturally acceptable salt or ester thereof, or a mixture thereof;

in association with one or more herbicidally-acceptable diluents or carriers and/or surface-active agents.

13. A herbicidal composition comprising a synergistic herbicidally effective amount of:

(a) a 4-benzoylisoxazole of formula (I):



(I)

wherein

R is hydrogen or $-\text{CO}_2\text{R}^3$;

R^1 is cyclopropyl;

R^2 is selected from halogen, $-\text{S}(\text{O})_p\text{Me}$ and C_{1-6} alkyl or haloalkyl;

n is two or three; p is zero, one or two; and

R^3 is C_{1-4} alkyl; and

(b) bromoxynil, which is 3,5-dibromo-4-hydroxybenzonitrile, or an agriculturally acceptable salt or ester thereof.

14. A composition according to claim 12 or 13 comprising from 1:120 to 16.7:1 wt/wt of (a) : (b) acid equivalent.

15. A product comprising a herbicidally effective amount of:

(a) a 4-benzoylisoxazole derivative of formula I as defined
5 in any one of claims 1, 2 or 6 to 9; and

(b) bromoxynil, which is 3,5-dibromo-
4-hydroxybenzonitrile, or ioxynil, which is 4-hydroxy-
3,5-diiodobenzonitrile, an agriculturally acceptable salt or ester
thereof, or a mixture thereof;

10 as a combined preparation for simultaneous, separate or
sequential use at a locus.

1/3

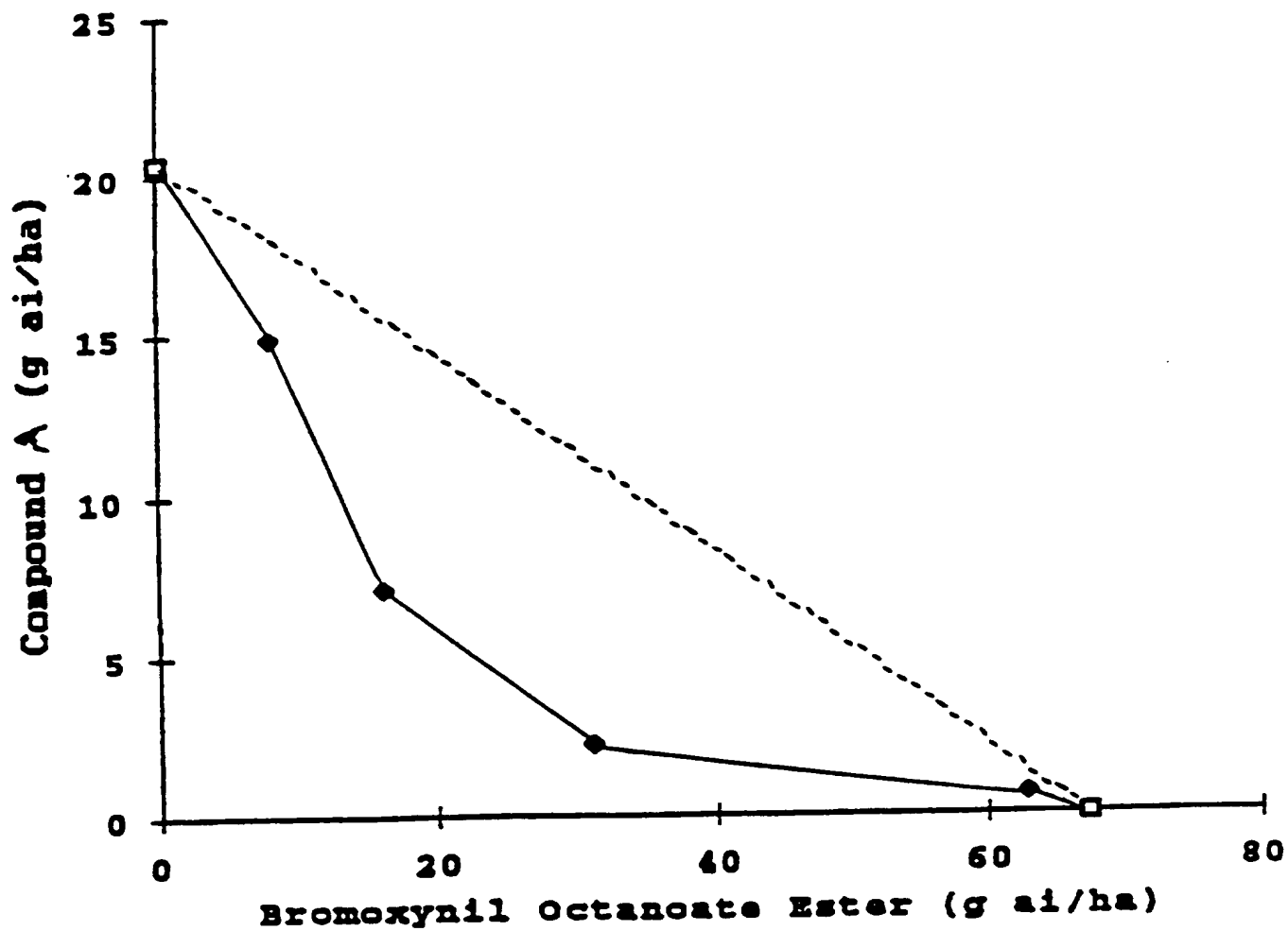


Fig.1

2/3

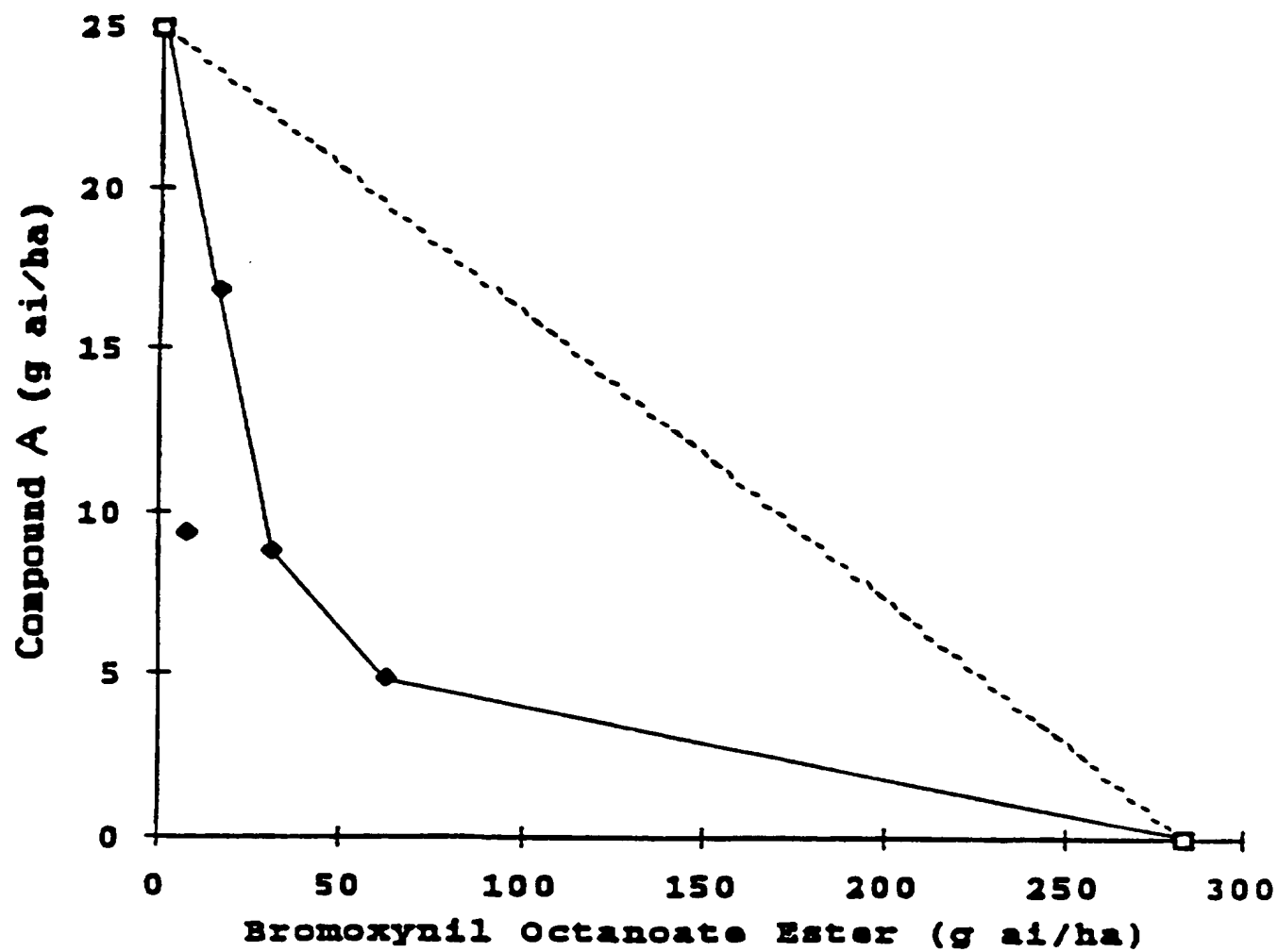


Fig.2

3/3

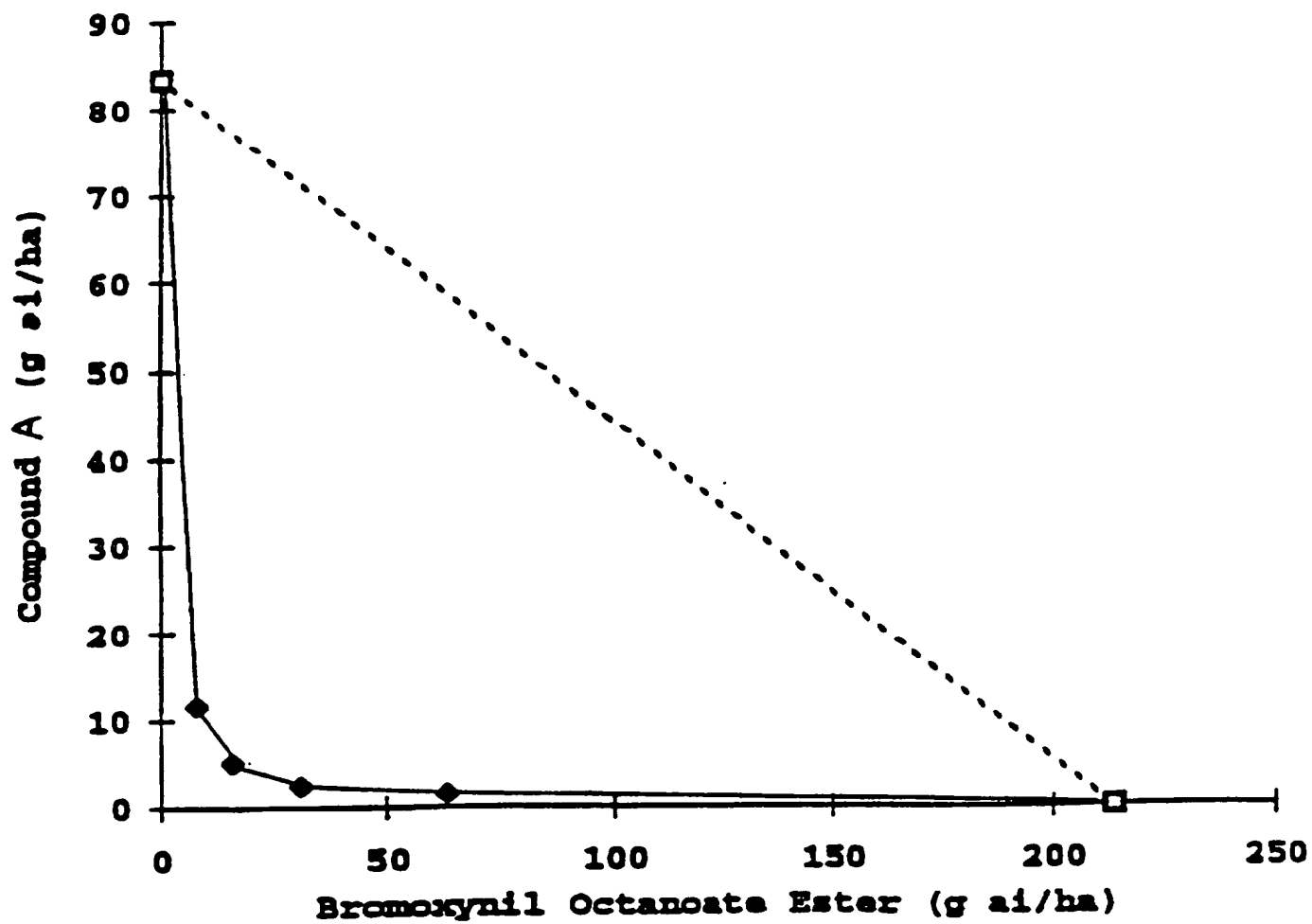


Fig. 3

INTERNATIONAL SEARCH REPORT

International Application No

PC., EP 96/05697

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 A01N43/80 //(A01N43/80,37:40)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 2 284 547 A (RHONE POULENC AGRICULTURE) 14 June 1995 see the whole document ---	1-15
A	FR 2 675 340 A (RHONE POULENC AGRICULTURE) 23 October 1992 see the whole document ---	1-15
A	CHEMICAL ABSTRACTS, vol. 110, no. 17, 24 April 1989 Columbus, Ohio, US; abstract no. 149835, XP002029870 see abstract & JP 63 216 806 A (HOKKO CHEMICAL INDUSTRY) 9 September 1988 --- -/--	1-15

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

- * "A" document defining the general state of the art which is not considered to be of particular relevance
- * "E" earlier document but published on or after the international filing date
- * "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- * "O" document referring to an oral disclosure, use, exhibition or other means
- * "P" document published prior to the international filing date but later than the priority date claimed

* "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

* "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

* "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

* "&" document member of the same patent family

Date of the actual completion of the international search

21 April 1997

Date of mailing of the international search report

- 7 -05- 1997

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl,
Fax (+ 31-70) 340-3016

Authorized officer

Lamers, W

INTERNATIONAL SEARCH REPORT

International Application No

PL./EP 96/05697

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 418 175 A (RHONE POULENC AGRICULTURE) 20 March 1991 cited in the application see page 34, line 58 see page 35, line 15 -----	1-15

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PL./EP 96/05697

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
GB 2284547 A	14-06-95	AU 1312295 A	27-06-95
		CN 1137221 A	04-12-96
		CZ 9601674 A	16-10-96
		WO 9515691 A	15-06-95
		EP 0732876 A	25-09-96
		PL 314961 A	30-09-96
		SK 74196 A	06-11-96

FR 2675340 A	23-10-92	CA 2065523 A	18-10-92
		CA 2065523 C	18-10-94
		IT 1254876 B	11-10-95
		US 5416061 A	16-05-95

EP 0418175 A	20-03-91	AT 140453 T	15-08-96
		AU 635316 B	18-03-93
		AU 6231390 A	14-03-91
		BG 60562 B	28-08-95
		CA 2024956 A	12-03-91
		CN 1050188 A	27-03-91
		CN 1141294 A	29-01-97
		DE 69027823 D	22-08-96
		DE 69027823 T	09-01-97
		EG 19315 A	29-02-96
		ES 2089003 T	01-10-96
		IL 95587 A	27-11-95
		JP 3118374 A	20-05-91
		OA 9311 A	15-09-92
		RU 2060663 C	27-05-96
		TR 25897 A	01-11-93

1/3

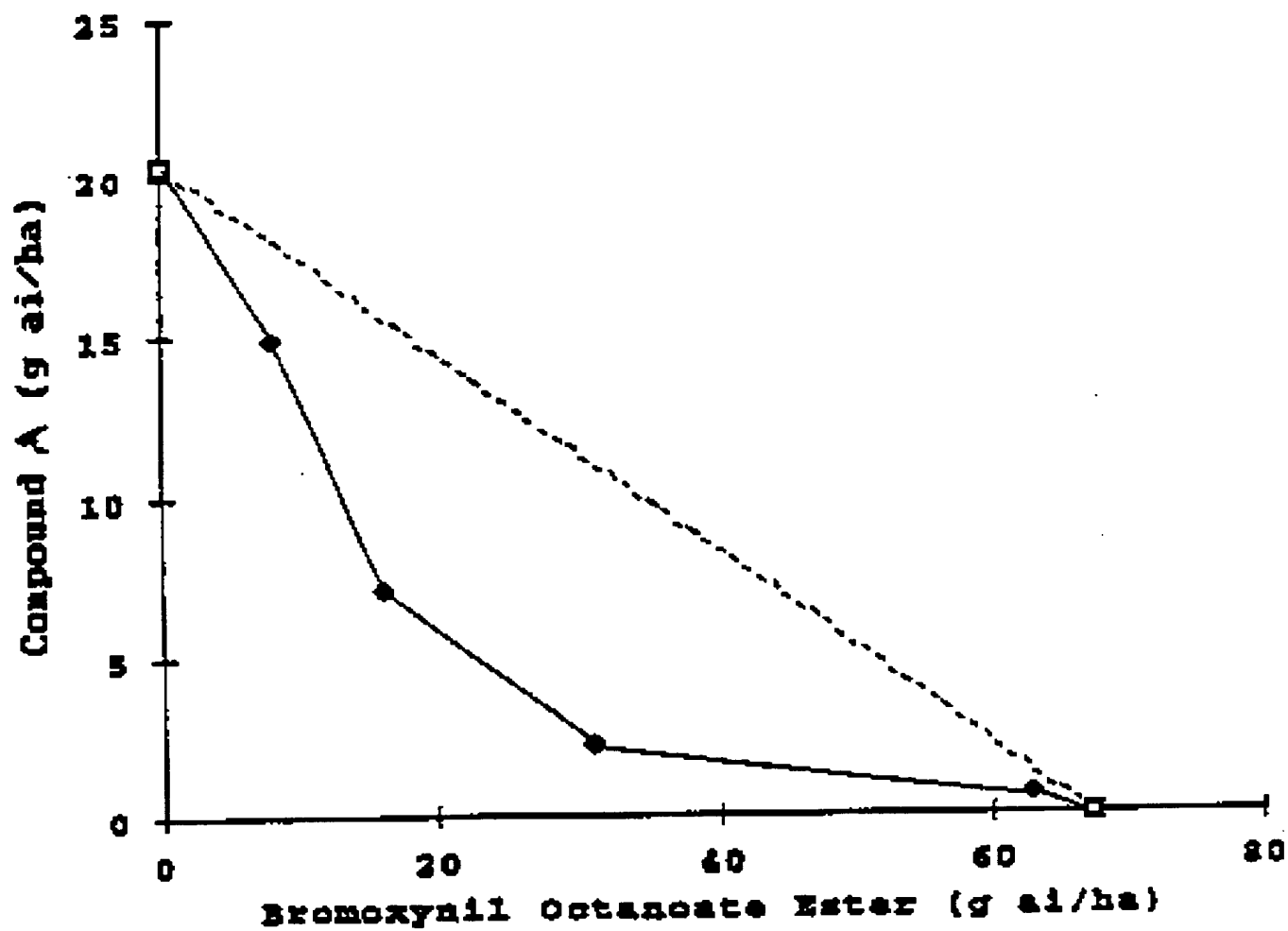


Fig. 1

2/3

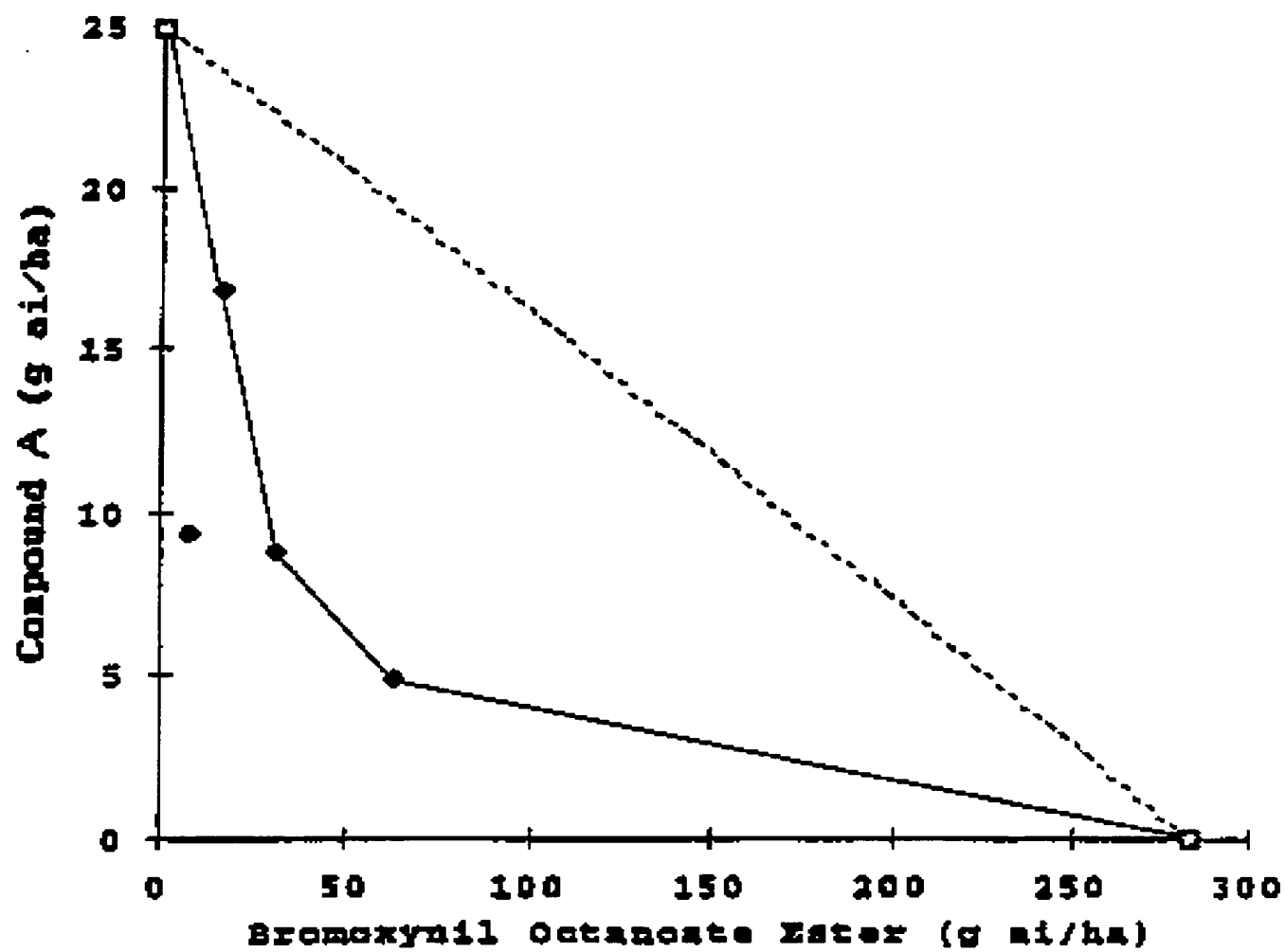


Fig.2

3/3

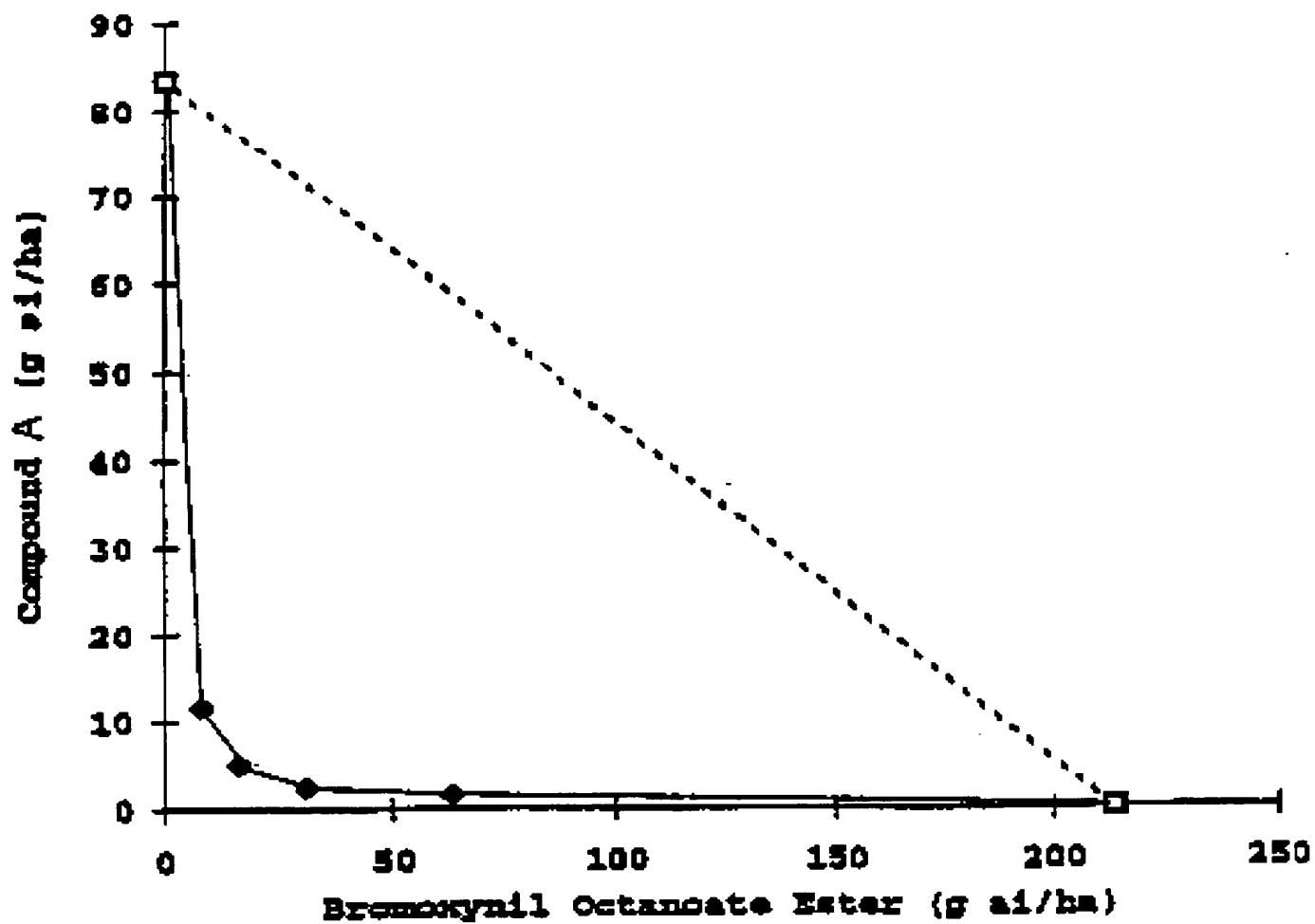


Fig. 3

THIS PAGE BLANK (USPTO)